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MODELING CREEP RUPTURE OF ZIRCONIUM ALLOYS

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ABSTRACT: Safe interim dry storage of spent nuclear fuel (SNF) must be maintained for a minimum of twenty years according to the Code of Federal Regulations. The most important variable that must be regulated by dry storage licensees in order to meet current safety standards is the temperature of the SNF. The two currently accepted models to define the maximum allowable storage temperature for SNF are based on a diffusion controlled cavity growth (DCCG) failure mechanism for the cladding. Although these models are based on the same fundamental failure theory (DCCG), the researchers who developed the models made different assumptions, including selection of some of the most critical variables in the DCCG failure equation. These inconsistencies are discussed together with recommended modifications to the failure models based on recent data.

INTRODUCTION: Interim dry storage of spent nuclear fuel (SNF) rods is of critical concern because of the shortage of wet storage capacity and delays in the availability of a permanent disposal site (mined geologic repository). The NRC has approved two models (Schwartz [1987]; Levy [1987]) to determine the maximum allowable temperature (MAT) for SNF in dry storage that supposedly meet all safety criteria and yield consistent temperature limits. Although these two models are based on the same fundamental failure theory, different assumptions have been made including the choice of values for material constants in the failure equation. This paper will discuss these differences as well as some of the shortcomings of the current models and suggest some modifications.

DISCUSSION: Currently, the MAT for dry storage of SNF is determined using either the equations developed by Lawrence Livermore National Laboratory (LLNL) (Schwartz [1987]) or temperature limit curves developed by Pacific Northwest National Laboratory (PNNL) (Levy [1987]). Both the PNNL and the LLNL models predict that cavitation failure under dry storage conditions may occur by diffusion controlled cavity growth (DCCG) (Raj [1975]). PNNL predicted temperature limits use a fracture map to account for various mechanisms predicted to be active over a relevant range of stresses and temperatures. For typical dry storage temperatures and stresses, the fracture map indicates that DCCG controls failure. Instead of DCCG, some have suggested using a 1% creep-strain limit approach (e.g., Peehs [1986]), which assumes that cavity growth and fracture only occur after significant plastic strain (>>1%). DCCG, however, is not strain dependent and it has been shown (Keusseyan [1979]) that cavities in zirconium alloy can nucleate and grow after very low plastic strains (much less than the strain to fracture).

PNNL uses a 'recovery factor' to account for the recovery of some of the reduction in ductility of zirconium alloys from irradiation damage. Because the DCCG fracture model is not a function of strain, a reduced ductility would not directly affect the fracture time and is not used by LLNL. PNNL assumes that failure occurs at an area fraction of decohesion of 0.50 (50%) whereas LLNL chose a value of 0.15 (15%) to define an upper decohesion limit. PNNL assumes a cavity spacing of 2.6 μm while LLNL assumes 10 μm.

Both models are very sensitive to changes in the temperature decay of the SNF (which has been modeled as following a power-type temperature decay) and the predicted MAT can be misleading because of this sensitivity, as illustrated in Fig. 1. Fig. 1 shows that the 40 and 5 year failure lines are coincident and very close to the 1 year failure line when using the power-decay temperature profile suggested by PNNL. Therefore an applicant who determines a MAT for a predicted failure of 300 years has, at the same time, predicted failure after 5 years. We evaluated how use of more conservative temperature decay profiles would affect predicted failure times based on the LLNL model (see Hayes [1999] for details). We found the sensitivity of the model is reduced with successively less concave (more linear) power-decay temperature profiles. This is because each successively less concave profile predicts more time at higher temperatures where the diffusion coefficients and stresses are higher and cavitation occurs more quickly. A linear profile is the most conservative. It predicts that more damage occurs (shorter fracture time) than any of the power decay profiles evaluated. It also decreases the sensitivity of the failure time to the initial temperature of the SNF compared to the temperature decay profile suggested by PNNL. Although measured profiles seem to obey power-like temperature decay, the uncertainty involved in developing the temperature profile (see discussion in Hayes [1999]) and the fact that the predicted failure time is so sensitive to this profile suggest using a conservative profile. We therefore recommend using an accurate temperature profile measured under the proposed storage configuration or, if unavailable, using a more conservative linear decay profile.

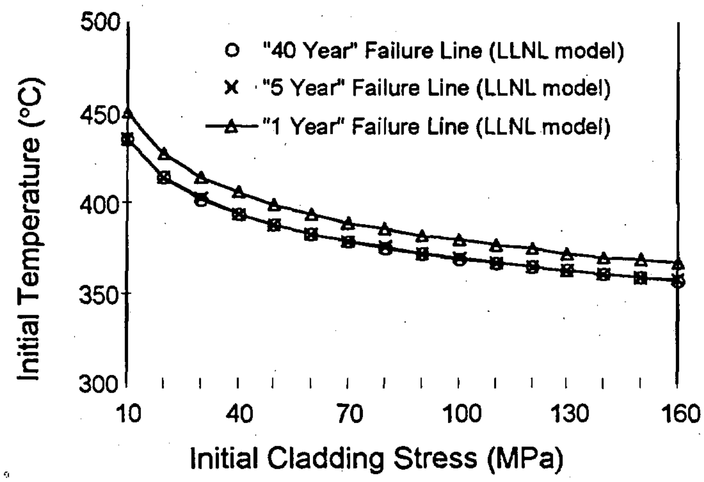

Figure 1. Comparison of 40, 5 and 1 year failure lines predicted using the LLNL model decay functions for 5-year-old SNF with an initial cladding hoop stress of 100 MPa.

Self-diffusion coefficients reported in the literature for zirconium (alloy data not reported) vary by more than four orders of magnitude. At the time these models were developed, no grain boundary diffusion studies had been reported for zirconium. The values chosen by LLNL and, presumably, by PNNL were based on an approximate relationship between the self-diffusion and grain boundary diffusion activation energies. Choosing a grain boundary diffusion coefficient based on these data, as PNNL and LLNL have done, yields diffusion coefficients that are 3 to 5 orders of magnitude different from one another. This is a serious limitation with these models as this variation translates linearly to the predicted failure time. Furthermore, it is not clear what effect irradiation may have on the grain boundary diffusion coefficient of zirconium alloys. Irradiation damage results in a higher concentration of vacancy type dislocation loops near grain boundaries and also causes dispersion of iron, which could substantially increase the diffusion rate. The effect of alloying on grain boundary diffusion under conditions relevant to dry storage has not been established, however zirconium alloys used for cladding of SNF are richer in iron (a fast diffuser in zirconium) than commercially pure zirconium (see Hayes [1999] for details) and thus may have a higher diffusion coefficient.

CONCLUSIONS: The fact that the two currently acceptable models are based on many inconsistent assumptions, combined with their extreme sensitivity to temperature leave the current models with questionable value. It should not be assumed that these models yield conservative temperature limits until experimental data for cavity spacing, grain boundary diffusion, and post-dry storage integrity of irradiated zirconium are obtained. A strain-based model is not applicable as long as DCCG is the assumed fracture mechanism.

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